

PATENT
Docket No.: 13199B

AFTER FINAL

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: :
: :
Wang et al. :
: :
Application No. 10/076,880 : Art Unit: 1754
: :
Filed: Feb. 13, 2002 : Examiner: E. Johnson
: :
For: REFORMING CATALYSTS AND : Conf. No. 8827
METHODS OF ALCOHOL STEAM :
REFORMING : Atty Docket: 13199B
: :
: :

BRIEF ON APPEAL

Board of Patent Appeals and Interferences
Commissioner for Patents
Washington, D.C. 20231

Dear Sir:

This Appeal Brief is submitted in accordance with the terms of 35 U.S.C. § 134 and 37 C.F.R. § 1.192 in response to the final Office Action mailed January 23, 2006. The \$250 small entity processing fee (37 C.F.R. § 41.20(b)(2)) is provided in documents accompanying this Brief; any fees required for consideration of this paper can be charged to deposit account 50-1749.

I. Real Party In Interest

Battelle Memorial Institute headquartered in Columbus, Ohio, is the real party in interest.

II. Related Appeals and Interferences

None.

III. Status of Claims

Claims 1, 5-26, and 31-35 are pending. Claims 2-4, and 27-30 have been canceled without prejudice or disclaimer. Claims 1, 5-23, 26 and 31-34 have been rejected and are the subject of this appeal.

IV. Status of Amendments

No amendments were filed subsequent to the final rejection.

V. Summary of the Invention

As can be seen from claims 1 and 5, the invention relates to catalyst comprising: a metal oxide support; a coating comprising zinc on the metal oxide support; and palladium in contact with the coating. The catalyst is further characterized by a very high volumetric productivity. The invention also includes a method of making a catalyst, in which a solution comprising dissolved zinc is combined with a solid metal oxide support. In this method, base is added to increase pH and, subsequent to at least a portion of the step of adding a base, depositing Pd.

VI. Grounds Of Rejection To Be Reviewed On Appeal

1. Are claims 1, 5-8, 9-15, 17-21 and 31-34 anticipated or rendered obvious by Wieland et al., U.S. Patent No. 6,413,449?
2. Are claims 16 and 22 unpatentable under 35 U.S.C. § 103 in view of the combination of Wieland et al., U.S. Patent No. 6,413,449 and Feinstein et al., U.S. Patent No. 4,177,219?

VII. Argument

I. Claims 1, 17, 18, 20, 21, and 23 Are Not Anticipated Or Rendered Obvious by the Wieland Patent Because the Catalyst Described in the Wieland Patent Does Not Possess the Claimed Volumetric Productivity.

Claim 1 recites that “the catalyst possesses a volumetric productivity of at least 10,000 ml H₂ / ml catalyst-hr.” Appellant’s specification defines “volumetric productivity” at page 3, lines 16-24. The conditions for measuring volumetric productivity are recited in claim 1.

The productivity value described in Wieland is provided in terms of liquid hourly space velocity and at conditions that do not conform with applicant’s definition of “volumetric productivity” at page 3, lines 16-24 which includes a specific protocol for measuring “volumetric productivity.” As explained in the Declaration (filed December 13, 2004) with attached document “Comparison of MeOH Steam Reforming Catalyst with U.S. Patent 6,413,449” (hereinafter both documents are referred to as “the 132 Declaration”), calculations show that Wieland’s catalyst does not inherently possess the claimed volumetric productivity.

The Examiner has stated that the Declaration is not persuasive because it “is specific to certain catalysts under certain process conditions, none of which are specified in the claim.” This statement is half-correct. It is correct in that the Declaration (specifically the “Comparison”) is specific to comparing Applicants’ claimed catalyst with the catalyst described by Wieland. It is incorrect in that the conditions for measuring volumetric productivity are specified in the claims.

The 132 Declaration (including the “Comparison”) prove that Wieland’s catalyst does not possess the claimed volumetric productivity. The Examiner does not dispute this fact.

The basis for patentability is simple. Appellants have claimed a catalyst that possesses a well-defined, measurable property – namely, “volumetric productivity.” Wieland’s catalyst does not possess this claimed feature. Therefore, Wieland does not anticipate or render obvious the invention of claim 1.

II. Catalyst Claims 19 and 20 and 26 are Additionally Patentable Over Wieland

Claims 19 and 20 recite higher levels of volumetric productivity and are therefore still further patentable over Wieland. The 132 Declaration proves that the Wieland catalyst does not possess these claimed levels of volumetric productivity.

III. Catalyst Claim 26 is Additionally Patentable Over Wieland

Claim 26 contains an additional limitation of specific activity. The 132 Declaration proves that the Wieland catalyst is less active than Applicants' catalyst. Therefore, the claimed level of specific activity is not inherent in Wieland's catalyst.

IV. The Invention of Claim 5 is Not Anticipated Or Rendered Obvious by Wieland Because Wieland Does Not Describe a Step of Adding a Solution Comprising Dissolved Zinc

The method of claim 5 is further patentable over Wieland et al., because Wieland et al. do not disclose a step of adding a solution comprising dissolved zinc to a metal oxide support, nor a step of adding a solid oxide support to a solution comprising dissolved zinc. Claim 5 requires "adding a solution comprising dissolved zinc to the solid metal oxide support, or adding a solid metal oxide support to a solution comprising dissolved zinc." In contrast, Wieland et al. combine a metal oxide with insoluble zinc oxide in an aqueous dispersion. It is true that Wieland et al. state that they subsequently dissolve a portion of the zinc in the acidic Pd solution; however, this does not constitute a step of "adding" a zinc solution. In other words, Wieland et al. state that they form a zinc solution *in situ*; however, they do not "add" a solution comprising dissolved zinc. Therefore, the rejection of claim 5 should be withdrawn since Wieland et al. do not "add" a solution comprising dissolved zinc, nor a "add" a metal oxide support to a solution comprising dissolved zinc.

On page 11 of the Office Action, the Examiner argues that " 'immersed' in the solution, is at least as specific as the claimed 'adding.' These are opposite processes. Clearly, there is no anticipation. Furthermore, in light of the well-known sensitivity of catalyst properties to the method by which they are made, it would not have been obvious that the opposite procedure of addition in place of the immersion step. Therefore, the method of claim 5 is not anticipated or rendered obvious by the Wieland reference.

V. Method of Making Claims 9, 12, and 32-34 are Additionally Patentable Over Wieland

Claim 9 recites that the zinc is completely dissolved in solution. In contrast, Wieland et al. do not teach any step of adding zinc that is completely in solution. Note that at col. 7, lines 33-40, Wieland et al. state that the finely divided zinc oxide powder is partially dissolved by the addition of the acidic noble metal solution. Although the Examiner responded to this argument

by referring to Example 1 of Wieland, Example 1 of Wieland lacks any step of adding a base. Therefore, Example 1 cannot anticipate the method of claim 9. Nor is there any motivation for modifying the procedure of Example 1 to add a base. Therefore, Wieland does not render obvious the method of claim 9.

Claim 12 recites a step of adding base after the zinc solution is added, while Wieland does not add any zinc solution. As discussed above, Wieland does not add a zinc solution. Furthermore, there is no motivation to modify Wieland's Example 1 by adding a base.

Claims 32-34 recite that the catalyst is prepared and reduced under hydrogen with temperatures never exceeding 400 °C. Maintaining low temperature increases catalyst life and surface area (see page 4, line 25 – page 5, line 1). All of Wieland's examples substantially exceed this temperature and, although broader ranges are mentioned, Wieland does not recognize the advantage of keeping process temperatures at 400 °C or less. Thus, claims 33-34 are patentable based on this additional ground.

VI. Calcining Does Not Constitute a Step of “Depositing Pd” Subsequent to Adding a Base. Therefore, the invention of claim 31 is not anticipated or rendered obvious by the Wieland Reference.

This argument is applicable to claim 31, and, to the extent that the Examiner maintains calcining as a step of depositing, also to claim 5.

In the Official Communication mailed January 25, 2005, the Examiner states that “The Pd of [Wieland] is considered deposited when the final product is produced by calcining.” This is erroneous. As is well-known, calcining involves heating a solid. For example, The Condensed Chemical Dictionary (9th Edition, 1977) defines calcination as “heating a solid to a temperature below its melting point to bring about a state of thermal decomposition or a phase transition other than melting.” Clearly, depositing Pd does not occur during a calcining step.

Claims are interpreted from the viewpoint of a person skilled in the art. The inventors (i.e., persons skilled in the art) stated in the 132 Declaration that:

Column 7, lines 13-17 in U.S. Patent No. 6,413,449 describe a part of a process of making a catalyst having the following steps “and is then reduced at constant temperature with an aqueous reducing agent, filtered, washed, dried, calcined in an oxidizing atmosphere at temperatures from 300°C to 550°C, and finally reduced in a hydrogen-containing gas at temperatures from 350°C to 500°C.” A

worker of ordinary skill in the field of making catalysts would not understand this part of the process as including any step of “depositing Pd.” There is no scientific basis to believe that Pd is deposited during the steps described in this excerpt.

In the example of Wieland, the catalyst powder was filtered and washed before it was dried and calcined. If any Pd remained in solution, it would be removed by filtering and washing, and therefore could not deposit during drying or calcining.

Unlike claim 31, Wieland does not teach or suggest a step of depositing Pd subsequent to a step of drying a zinc-containing layer.

VI. Claims 16 and 22 are Patentable Over the combination of Wieland and Feinstein et al., U.S. Patent No. 4,177,219.

A. The Feinstein Patent is Not Analogous Art.

Applicant’s catalyst and Wieland’s catalyst relate to methanol steam reforming:



On the other hand, Feinstein relates to a method for converting (reforming¹) ethyl aromatics to methyl aromatics (i.e., xylenes), for example:



The standard for analogous art is set forth in MPEP 2141.01(a) (“In order to rely on a reference as a basis for rejection of an applicant’s invention, the invention must either be in the field of applicant’s endeavor or, if not, then be reasonably pertinent to the particular problem with which the inventor was concerned.”) See also *In re Clay*, 23 USPQ2d 1058 (Fed. Cir. 1992).

The reforming of ethyl aromatics to xylenes is not in the same field as reforming methanol to hydrogen and carbon dioxide. The chemistries are entirely different. As can be seen from the chemical equations above, one process relates to the production of H₂ while the other process relates to the production of xylenes. Thus, Feinstein’s process of converting ethyl aromatics to xylenes is not analogous art.

¹ The only common feature shared by appellant’s claimed invention and the Feinstein patent is the term “reforming.” However, the reforming processes are completely different. Martin Luther attempted to reform the Catholic Church, and the juvenile justice system attempts to reform wayward youth – both of these use the term “reforming,” but they too are nonanalogous art to methanol steam “reforming.”

Nor is Feinstein's process "reasonably pertinent to the particular problem with which the inventor was concerned." The problem with which the inventors were concerned was the reaction of methanol with water to produce hydrogen and carbon dioxide. The problem Feinstein was concerned with was the scission of the C-C bond in the ethyl group in ethyl aromatics; this is a completely different problem. Unlike ethyl aromatics, methanol does not have any C-C bonds. Thus, catalyst compositions for the conversion of ethyl aromatics to xylenes are not reasonably pertinent to the problem with which the inventors were concerned.

Therefore, the Feinstein patent is nonanalogous art and it cannot be used in a section 103 rejection.

B. There is Not a Proper Motivation to Combine the Wieland and Feinstein patents.

The Examiner is in error in stating that "high scission activity" of Feinstein's catalyst provides a motivation to add Ru into Wieland's catalyst. In Feinstein's process reforming ethyl aromatics to produce methyl aromatics, the scission of the C-C bond in the ethyl group is critical to forming methyl aromatics. In contrast, methanol does not have any C-C bonds. Therefore, the "high scission activity" of Feinstein's catalyst is irrelevant to methanol steam reforming.

The situation is similar with regard to conversion and selectivity. Feinstein's catalyst demonstrated good conversion of ethyl aromatics. There are no ethyl aromatics in methanol steam reforming. The selectivity that Feinstein refers to is selectivity to xylenes. In contrast, methanol steam reforming produces no xylenes. With or without Ru, the selectivity to xylenes will be zero in methanol steam reforming. A worker of ordinary skill in the field of methanol steam reforming would know that the catalysts and mechanisms for converting ethyl aromatics to methyl aromatics are not relevant to the catalysts and mechanisms involved in methanol steam reforming (this is stated in the 132 Declaration). Therefore, there is not a proper motivation to combine the teachings of the Wieland and Feinstein references.

Conclusion

For the foregoing reasons, appellants respectfully submit that the Examiner has erred in rejecting this application. Please reverse the Examiner on all counts.

Dated this 21st day of June, 2006.

Respectfully submitted,

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VII. CLAIMS APPENDIX

1. A catalyst comprising:
a metal oxide support;
a coating comprising zinc on the metal oxide support; and
palladium in contact with said coating;
wherein the catalyst possesses a volumetric productivity
such that, when tested in a measurement in which the catalyst is placed in a reactor and exposed
to premixed, vaporized water and methanol at a water to methanol ratio of 1.78, at 300 °C, about
1 atm pressure and a contact time of 100 ms (for powders and crushed pellets) or 150 ms (for
felts and foams), at least 10,000 ml H₂ / ml catalyst·hr is produced.
2. (canceled)
3. (canceled)
4. (canceled)
5. A method of making a catalyst, comprising the steps of:
providing a solid metal oxide support;
adding a solution comprising dissolved zinc to the solid metal oxide support, or adding a
solid metal oxide support to a solution comprising dissolved zinc;
adding a base to increase pH; and
subsequent to at least a portion of the step of adding a base, depositing Pd.
6. A catalyst made by the method of claim 5.
7. The method of claim 5 wherein the metal oxide support comprises alumina, titania or
zirconia.
8. The method of claim 5 wherein the metal oxide support is deposited onto a large pore
support.

9. The method of claim 5 comprising a step wherein the zinc is completely dissolved in said solution.
10. The method of claim 5 wherein there are no metals other than zinc in said solution.
11. The method of claim 5 wherein said solution comprises 0.1 to 3 M zinc.
12. The method of claim 5 wherein the base is added after the zinc solution is added.
13. The method of claim 12 wherein base is added to result in a pH of 7 or greater.
14. The method of claim 13 further comprising a step of calcining at 200 to 400 °C.
15. The method of claim 12 wherein Pd is deposited from a solution comprising dissolved Pd.
16. The method of claim 15 wherein the solution comprising Pd further comprises Ru.
17. The catalyst of claim 1 wherein the metal oxide support constitutes 50 to 90 wt% of the catalyst; zinc oxide constitutes 10 to 30 wt% of the catalyst; and Pd constitutes 1 to 15 wt% of the catalyst.
18. The catalyst of claim 17 wherein the metal oxide support comprises alumina, titania or zirconia.
19. The catalyst of claim 17 possessing a volumetric productivity of at least 40,000 ml H₂/ml catalyst·hr.
20. The catalyst of claim 17 possessing a volumetric productivity of 20,000 to 90,000 ml H₂/ml catalyst·hr.

21. The catalyst of claim 20 comprising 2 to 10 wt% Pd.
22. The catalyst of claim 21 comprising 0.2 to 5 wt% Ru.
23. The catalyst of claim 1 wherein the metal oxide forms a layer having a thickness less than 1 mm on a large pore support.
24. The catalyst of claim 23 wherein the metal oxide forms a layer having a thickness less than 40 μm , and the large pore support comprises a foam or felt.
25. The catalyst of claim 23 wherein at least 50% of the catalyst's pore volume is composed of pores in the size range of 0.3 to 200 microns.
26. The catalyst of claim 17 characterizable by a specific activity of greater than 1.5 mol methanol converted/(g catalyst)(hr) when tested at 400 $^{\circ}\text{C}$, 25 msec contact time, 1.8 steam-to-carbon ratio with a pressure drop of less than 25 psig.

27-30. (Canceled)

31. The method of claim 5 wherein a zinc-containing layer is formed on the metal oxide support; and
 32. further comprising a step of drying the zinc-containing layer; wherein the step of depositing Pd occurs subsequent to said step of drying.
33. The method of claim 31 wherein the catalyst is prepared and reduced under hydrogen with temperatures never exceeding 400 $^{\circ}\text{C}$.
34. The method of claim 15 wherein the catalyst is prepared and reduced under hydrogen with temperatures never exceeding 400 $^{\circ}\text{C}$.

34. The method of claim 5 wherein the catalyst is prepared and reduced under hydrogen with temperatures never exceeding 400 °C.

35. The method of claim 5 wherein Pd is deposited by impregnation from Pd dissolved in solution.

VIII. EVIDENCE APPENDIX

1. Copy of Declaration Pursuant to 37 C.F.R. § 1.132 signed February 10, 2004 and filed December 13, 2004.
2. Comparison of MeOH Steam Reforming Catalyst with U.S. Patent 6,413,449 submitted along with the section 132 Declaration referred to above.